

Energy transfer and upconversion in Tm^{3+} -doped $\beta\text{-NaYF}_4$: comparison between models and theory

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Tm^{3+} ions in close proximity interact non-radiatively in different ways so that, for example, blue $^3\text{H}_6 \rightarrow ^1\text{G}_4$ excitation (473 nm) results in UV $^1\text{D}_2 \rightarrow ^3\text{H}_6$ upconverted emission (361 nm) in a $\beta\text{-NaYF}_4$: 0.3% Tm^{3+} sample, see Fig 1.

The strength of an energy transfer (ET) interaction can be summarized by a single number: the critical radius R_c . This is the distance at which the probability of ET and radiative decay are equal. At shorter distances ET dominates, while at longer distances radiative decay is more important.

A recent model is able to obtain the R_c of different interactions by fitting luminescence decay curves, see Fig 1(b). [1] For the cross-relaxation interaction shown in Fig. 1(a), this model determines $R_c = 11.8 \text{ \AA}$. An analysis based on the Inokuti-Hirayama model results in $R_c = 12.1 \text{ \AA}$.

An equation due to T. Kushida determines R_c as a function of the donor state lifetime, refractive index, transition energy, emission and absorption oscillator strengths, and spectral overlap. [2]

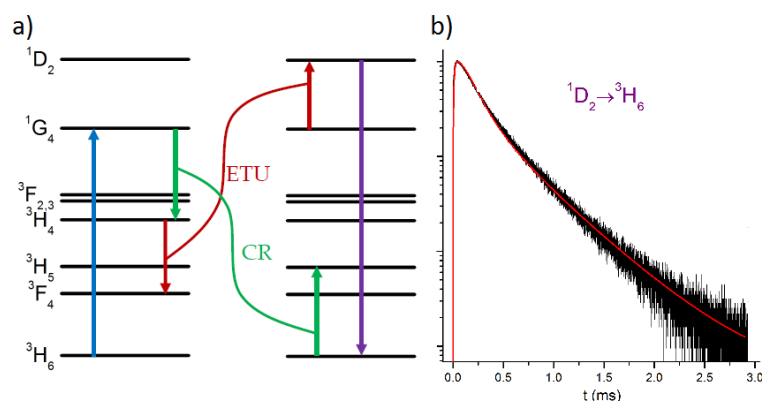


Fig. 1. a) Electronic structure of Tm^{3+} and relevant absorption, emission, and energy transfer processes in $\beta\text{-NaYF}_4$: 0.3% Tm^{3+} . b) $^1\text{D}_2 \rightarrow ^3\text{H}_6$ luminescence decay curve (361 nm) after $^3\text{H}_6 \rightarrow ^1\text{G}_4$ excitation (473 nm) with model fit (red line).

These parameters have been determined for a $\beta\text{-NaYF}_4$: 0.3% Tm^{3+} powder sample and a $\beta\text{-NaGdF}_4$: Yb^{3+} , Tm^{3+} single crystal. [1,3] The predicted critical radius is $R_c = 7.6 \text{ \AA}$. The disagreement between the model and theory can be explained by the difficulty in measuring the spectral overlap, due to the disorder in the $\beta\text{-NaYF}_4$ lattice.

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